## Magnetic relaxation and the lower critical fields in a Y-Ba-Cu-O crystal

Y. Yeshurun, \* A. P. Malozemoff, F. Holtzberg, and T. R. Dinger IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598 (Received 17 June 1988)

We report a low-temperature study of magnetic relaxation for fields 30 Oe  $\leq H \leq$  30 kOe applied along the principal axes of a Y-Ba-Cu-O crystal. The lower critical fields are identified by the onset of relaxation at 900  $\pm$  100 Oe and 250  $\pm$  50 Oe for H||c and H $\perp$ c, respectively. The field dependence of the relaxation rates above the critical fields is interpreted with a thermally activated flux creep model.

The characterization of the new high- $T_c$  superconductors would not be complete without an accurate determination of the critical fields. Yet the values for both the lower  $(H_{c1})$  and the upper  $(H_{c2})$  critical fields are still controversial. First reports on the values of the critical fields in polycrystalline materials had inevitable averaging of the anisotropic crystalline values and possible glassy contributions which mask the bulk features. It is therefore natural to expect that single-crystal studies would properly identify the critical fields. So far, however, upper-critical-field determinations have been masked by irreversible effects, while reported lower critical fields  $H_{c1}^{\parallel}$  and  $H_{c1}^{\perp}$  range from 690 Oe-5 kOe for H || c and from 70 Oe-2 kOe for  $H \perp c$ , respectively. <sup>3-6</sup> This large span of values reflects the experimental difficulty in determining the threshold for deviations from linearity in conventional magnetization versus field curves, especially in the presence of strong pinning.

In the present article we report a new method for determining  $H_{c1}$ , using the onset of irreversibility above this field, and following an early suggestion of Worthington, Gallagher, and Dinger. For fields  $H \leq H_{c1}$  flux is completely expelled and the magnetization  $(M = -H/4\pi)$  is stable. Only above  $H_{c1}$  does flux penetrate into the sample to form the metastable mixed state. Metastability is borne out in experiment by observation of a logarithmic decay of the magnetization. However, the onset of relaxation at  $H_{c1}$  turns out not to be sharp. To properly determine  $H_{c1}$ , we extend the usual flux-creep and critical-state models, which permits a fit of the relaxation field dependence. We argue that this method is considerably more reliable than conventional field-dependent magnetization measurements at low temperatures and it complements a higher-temperature method to be reported elsewhere.

Relaxation measurements have been performed on

three crystals grown by different techniques.  $^{13,14}$  Table I summarizes the relevant information for the three crystals. We present here low-temperature (6 K) data for sample No. 2 of Table I, a  $1000 \times 480 \times 25 \ \mu\text{m}^3$  crystal with  $T_c$  =91.5 K. The orthorhombic c axis (perpendicular to the CuO planes) is along the shortest edge. The temperature dependence of the relaxation for this sample is presented in Ref. 15 and it has the same qualitative features as for another sample, given earlier in Ref. 16. The magnetic measurements have been performed on a commercial SHE superconducting quantum interference device (SQUID) magnetometer using a conventional procedure: The sample is zero-field cooled (zfc). At 6 K a field H (30 Oe  $\leq H \leq$  30 kOe) is applied and the magnetization is measured for typically 1 h.

Figures 1 and 2 summarize the field dependence of the logarithmic relaxation rates  $S \equiv dM/d \ln t$  determined at 6 K for both orientations. The field is corrected for demagnetization using an ellipsoidal approximation to the sample shape. (In the low-field limit this correction yields a susceptibility value which is within 90% of  $-1/4\pi$ .) In both figures S increases with field, reaches a maximum at a field  $H_m$  and then slows down. The insets to Figs. 1 and 2, in which  $S/H^2$  is plotted versus H demonstrate that below  $H_m$  but above a threshold  $H_{c1}$ , S increases as  $H^3$  for both  $H \parallel c$  and  $H \perp c$ . Such dependences of relaxation on H have earlier been reported on ceramic materials by Mota et al. 17

Logarithmic time dependences of the magnetization are well documented in the literature for low  $T_c$  (Refs. 8-10) as well as for the new high- $T_c$  superconductors. <sup>15-17</sup> To explain the logarithmic relaxation, Anderson suggested a flux creep model in which flux lines in the critical state hop over potential barriers  $U_0$  due to thermal activation. To interpret the present results we extend the conventional flux creep model <sup>7-10</sup> to include field dependence. In

TABLE I. Summary of the information for the three samples of this experiment.  $N^{\parallel}$  and  $N^{\perp}$  are the demagnetization correction factors for  $H \parallel c$  and  $H \perp c$ , respectively.  $H_{c1}$  and  $U_0$  are the results of the fit of S vs H data to Eq. (6).

Sample	Ref.	$T_c$ (K)	$N^{\parallel}$	N <sup>⊥</sup>	$H_{c1}^{\parallel}$ (Oe)	$H_{c1}^{\perp}$ (Oe)	$U_0^{\parallel}$ (eV)	$U_0^{\perp}$ (eV)
1	13	85	0.7	0.15	900	240	0.018	0.4
2	14	91.5	0.94	0.03	950	230	0.02	0.15
3	14	90.5	0.64	0.18	800	280	0.06	0.5

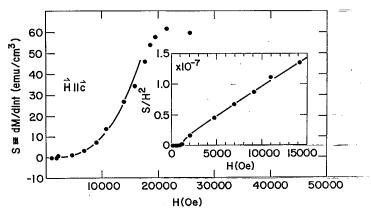


FIG. 1. Relaxation rate of the zero-field-cooled magnetization as a function of field (corrected for demagnetization) for fields parallel to the orthorhombic c axis of an YBaCuO crystal. Inset: relaxation rate  $S = dM/d \ln t$  normalized by  $H^2$  as a function of field. Solid lines are fit to Eq. (6) with n = 1. Broken line is a guide for the eye.

the following we first derive the field dependence of the low-temperature magnetization; then we incorporate the flux creep effect to derive the field dependence of the relaxation rate.

We develop a formula for the magnetization of a slab of thickness D with field in the slab plane. Extending the Bean critical state model<sup>11</sup> we take  $J_c$  to be field dependent:

$$J_c = J_{c1}(H_{c1}/h)^n, \quad h > H_{c1}, \tag{1}$$

where  $J_{c1}$  is the maximum critical current at a given temperature, h is the local field at a location x relative to the edge of the slab, and n is a phenomenological power, typically 1 in our measurements. In a critical state  $dh/dx = -4\pi J_c/10$ . (Here and throughout this paper we use practical units, i.e., Oe, cm, and A/cm<sup>2</sup>.) If we assume h(x) equals the applied field H at the surface boundary

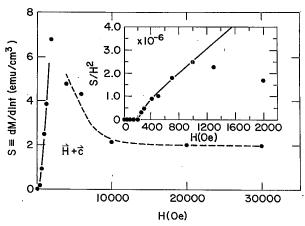


FIG. 2. Relaxation rate of the zero-field-cooled magnetization as a function of field (corrected for demagnetization) for fields perpendicular to the orthorhombic c axis of an YBaCuO crystal. Inset: relaxation rate  $S = dM/d \ln t$  normalized by  $H^2$  as a function of field. Solid lines are fit to Eq. (6) with n=1. Broken line is a guide for the eye.

x = 0, the contour of the local fields is given by

$$h^{n+1} = (H^{n+1} - Cx), \quad C = \frac{4\pi}{10} (n+1) J_{c1} H_{c1}^n.$$
 (2)

Figure 3 sketches the local fields h vs x for a field  $H \le H^*$  where  $H^* \equiv (CD/2 + H_{c1}^{n+1})^{1/(n+1)}$  is the first field for which currents flow through the entire volume of the sample, i.e.,  $x_c = D/2$ . We assume here that h(x) drops to zero at  $H_{c1}$ . This is a simple approximation to the "dB/dH effect" discussed in the literature; <sup>18,19</sup> a more general treatment of this effect, to be presented elsewhere, does not alter the main conclusions of this work.

Since the magnetization  $4\pi M$  is given by  $\overline{B} - M$  with  $\overline{B}$  a spatial average over h(x),  $H + 4\pi M$  is zero below  $H_{c1}$  and

$$H + 4\pi M = \frac{2}{CD} \frac{n+1}{n+2} (H^{n+2} - H_{c1}^{n+2}), \quad H_{c1} \le H \le H^*,$$
(3)

$$H + 4\pi M = \frac{2}{CD} \frac{n+1}{n+2} \left[ H^{n+2} - \left( H^{n+1} - \frac{CD}{2} \right)^{(n+2)/(n+1)} \right], \quad H \ge H^*.$$
 (4)

The time dependence in Eqs. (3) and (4) is implicit in the quantity C which depends on  $J_c$ . Flux creep results in a time-dependent reduction in critical current which at low temperature is described by  $^{10}$ 

$$J = J_{c0} \left[ 1 - \frac{kT}{U_0} \ln \frac{t}{t_0} \right], \tag{5}$$

where  $J_{c0}$  is the critical current in the absence of thermal activation, t is the measurement time, and  $1/t_0$  is a characteristic attempt frequency for flux hopping over pinning barriers, typically<sup>2</sup> of order  $10^9$  Hz. Inserting Eq. (5) into Eqs. (3) and (4), we find no relaxation below  $H_{c1}$ 

and to lowest order in  $kT/U_0$ 

$$\frac{dM}{d\ln t} = \frac{1}{4\pi} \frac{2}{C_0 D} \frac{n+1}{n+2} (H^{n+2} - H_{c1}^{n+2}) \frac{kT}{U_0} , \qquad (6)$$

 $H_{c1} \leq H \leq H^*$ ,

$$\frac{dM}{d\ln t} = \frac{1}{4\pi} \frac{1}{2(n+1)} \frac{C_0 D}{2} H^{-n} \frac{kT}{U_0}, \quad H \gg H^*, \tag{7}$$

where  $C_0$  is the zero-temperature value of C, i.e., in the absence of thermal activation. Note that in the low-field limit Eq. (6) also describes the relaxation of a cylinder of radius R, provided D is replaced by R in Eq. (6) and D/2 is replaced by R in the expression for  $H^*$ . High-

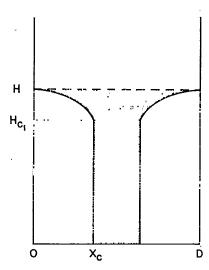


FIG. 3. Schematic position dependence of local fields in a slab of thickness d for a field  $H \leq H^*$ .

temperature corrections for Eq. (7) (in the limit  $kT/U_0 \gg 1$ ) are discussed in Ref. 20.

Qualitatively, the rise and fall of  $dM/d \ln t$  with field shown in Figs. 1 and 2 are predicted by Eqs. (6) and (7), respectively. In particular, Eq. (7) which predicts a decrease in the relaxation rate accounts qualitatively for the sharp peak observed here (Fig. 2) and in conventional superconductors. 9 Equation (7) predicts however too fast a decrease in  $dM/d \ln t$  at higher fields ( $\simeq H^{-2}$  dependence for n = 1, taking into account the implicit field dependence in  $C_0$ ). We note however that at high temperatures and high fields the logarithmic decay is no longer a good approximation of the experimental behavior. In fact, in this regime we observe 16 strong deviations from logarithmic decay. The dM/d lnt values reported here in the highfield limit are derived at the end of the time window of the measurement. Slower logarithmic relaxation is expected at much larger time scales.

In the low-temperature low-field limit where the logarithmic time decay is a good approximation of the experimental situation, Eq. (6) accounts for the observed data. The insets to Figs. 1 and 2 demonstrate that in this limit Eq. (6) describes the data best with exponent 3 (i.e., n=1). The solid lines in these figures and in the respective insets are fits to Eq. (6) for the low-field data with n=1 and threshold for detectable magnetic relaxation at  $H_{c1}^{\parallel}$  =950 Oe and  $H_{c1}^{\perp}$  =230 Oe. These  $H_{c1}$  values are summarized in Table I together with the results of a similar study on two other crystals. The most remarkable point in this table is that despite the diversity in demagnetization factors and the different  $U_0$  values, the scatter in  $H_{c1}$  is modest and of the same order as the magneticfield step in this experiment. Averaging the results in Table I, we estimate  $H_{c1}^{\parallel} = 900 \pm 100$  Oe and  $H_{c1}^{\perp}$  $=250\pm50$  Oe, values to be discussed further below. These results are smaller than those reported in Refs. 3 and 4 and are consistent with the results of the torque experiments in Ref. 6.

From the range of validity of Eq. (6), demonstrated in the insets of Figs. 1 and 2, we estimate  $H_{\parallel}^* \approx 14$  kOe and

 $H_{-}^{**}=1$  kOe for H||c and H $_{-}$ c, respectively. From the definition of  $H^{*}$  and with the given sample dimensions we find the zero-temperature critical currents. For H||c with D/2=0.024 cm, n=1, and  $H_{c1}=950$  Oe we find  $J_{c0}^{\parallel}=3\times10^{6}$  A/cm<sup>2</sup>. Similarly for H $_{-}$ c with D=0.0025 cm, n=1, and  $H_{c1}=230$  Oe,  $J_{c0}^{\perp}=1\times10^{6}$  A/cm<sup>2</sup>. It is important to note that with these parameters  $J_{c0}$  depends only weakly on  $H_{c1}$ . The absolute  $J_{c0}$  values derived here as well as the anisotropy ratio are not far from published data, e.g., see Ref. 3. The only parameter left unknown is the potential barrier  $U_{0}$ . From the derived  $J_{c0}$  values and the high-field slope in the fits of Figs. 1 and 2, we finally determine  $U_{0}^{\parallel}=0.02$  eV and  $U_{0}^{\perp}=0.15$  eV.

In previous papers  $^{15,16}$  we derived  $U_{0}$  values from the

In previous papers <sup>15,16</sup> we derived  $U_0$  values from the temperature dependence of  $dM/d \ln t$ . The values derived here from the field dependence of  $dM/d \ln t$  confirm our main conclusion, namely that  $U_0$  is unusually small compared to conventional type-II superconductors. (In Ref. 16 we used the conventional formula which assumes  $H \ge H^*$  for the temperature dependence of the relaxation. The more appropriate treatment at low-temperature, considering that  $H < H^*$ , yields similar  $U_0$  values as obtained here). As pointed out already, <sup>16,21</sup> the small  $U_0$  values might be a direct consequence of the small coherence lengths which characterize the new high  $T_c$  superconductors.

Conventionally  $H_{c1}$  is determined from the onset of nonlinearity in M vs H curves. By comparing Eqs. (4) and (6), it is apparent that the nonlinearity has the same field dependence as the relaxation rate. However, in the conventional measurements the large linear "baseline" masks the relatively small deviations; this problem may explain the large span of reported values<sup>3-5</sup> for  $H_{c1}$ . To demonstrate the experimental difficulties in determining deviations from linearity, it should be noted<sup>6</sup> that according to Ref. 5 the relative deviation at  $3H_{c1}$  is only of order 1% of the initial  $1/4\pi$  slope. Thus relaxation measurements are advantageous in that they bypass the need for a background subtraction. Furthermore, because of the relatively large relaxation effects a conventional measurement represents a series of data points at ever longer times whereas the relaxation measurement, while longer to perform explicitly takes into account the time dependence of the data. It is nevertheless noteworthy that the raw data of Figs. 1 and 2 curve upward in a smooth way; so that it is difficult to pick out an  $H_{c1}$  value. Our theory thus provides the needed method for properly extrapolating the data to determine  $H_{c1}$ , as illustrated by the plots in the insets to Figs. 1 and 2.

Now that the earlier measurements of  $H_{c2}$  are under question,<sup>2</sup> these measurements of  $H_{c1}$  provide perhaps the most reliable measure of anisotropy in the superconducting properties. The field anisotropy is only a factor of 4, considerably lower than previously thought. The values of  $H_{c1}$  imply anisotropic low-temperature penetration depths of 1000 and 4600 Å, according to classic formulas.<sup>22</sup>

The authors thank T. Worthington, R. Greene, L. Krusin-Elbaum, J. R. Clem, D. Johnston, and V. Kogan for helpful conversations.

- \*Permanent address: Department of Physics, Bar-Ilan University, Ramat-Gan, Israel.
- <sup>1</sup>S. Senoussi, M. Oussena, and S. Hadjoudj, J. Appl. Phys. 63, 4176 (1988).
- <sup>2</sup>A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. H. Kes, Phys. Rev. B 38, 7203 (1988).
- <sup>3</sup>T. K. Worthington, W. J. Gallagher, and T. R. Dinger, Phys. Rev. Lett. **59**, 1160 (1987); W. J. Gallagher, T. K. Worthington, T. R. Dinger, F. Holtzberg, D. L. Kaiser, and R. L. Sandstrom, Physica B **148**, 228 (1987).
- <sup>4</sup>T. R. McGuire, T. R. Dinger, P. J. P. Freitas, W. J. Gallagher, T. S. Plaskett, R. L. Sandstrom, and T. M. Shaw, Phys. Rev. B 36, 4032 (1987); T. R. McGuire, F. Holtzberg, D. L. Kaiser, T. M. Shaw, and S. Shinde (unpublished).
- <sup>5</sup>A. Umezawa, G. W. Crabtree, J. Z. Liu, T. J. Moran, S. K. Malik, L. H. Nunez, W. L. Kwok, and C. H. Sowers, Phys. Rev. B 38, 2843 (1988).
- <sup>6</sup>L. Fruchter, C. Giovannella, G. Collin, and I. A. Campbell, Physica C 156, 69 (1988).
- <sup>7</sup>P. W. Anderson, Phys. Rev. Lett. 9, 309 (1962).
- 8P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. 36, 39 (1964).
- <sup>9</sup>M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. 181, 682 (1969).
- <sup>10</sup>A. M. Campbell and J. E. Evetts, Adv. Phys. 21, 199 (1972).

- <sup>11</sup>C. P. Bean, Phys. Rev. Lett. 8, 250 (1962); Rev. Mod. Phys. 36, 31 (1964).
- <sup>12</sup>L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronemeyer, and F. Holtzberg (unpublished).
- <sup>13</sup>T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. 58, 2687 (1987).
- <sup>14</sup>D. L. Kaiser, F. Holtzberg, M. F. Chisholm, and T. K. Worthington, J. Cryst. Growth 85, 593 (1987).
- 15Y. Yeshurun, A. P. Malozemoff, and F. Holzberg, J. Appl. Phys. (to be published).
- <sup>16</sup>Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
- <sup>17</sup>A. C. Mota, A. Pollini, P. Visani, K. A. Müller, and J. G. Bednorz, Phys. Rev. B 36, 401 (1987).
- <sup>18</sup>J. R. Clem, J. Appl. Phys. **50**, 3518 (1979).
- <sup>19</sup>H. Ullmaier, Irreversible Properties of Type II Superconductors, Springer Tracts in Modern Physics, Vol. 76 (Springer-Verlag, New York, 1975).
- <sup>20</sup>A. P. Malozemoff, T. K. Worthington, R. M. Yandrofski, and Y. Yeshurun, in *Towards the Theoretical Understanding of High-T<sub>c</sub> Superconductors*, edited by S. Lundqvist, E. Tosatti, and Y. Lu (World Scientific, Singapore, in press).
- <sup>21</sup>M. Tinkham, Helv. Phys. Acta **61**, 443 (1988).
- <sup>22</sup>V. Kogan, Phys. Rev. B 24, 1572 (1981).